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Radiological risk from thoron, a case study: The particularly radon-prone area of Bolsena, and the lesson learned

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HIGHLIGHTS

- 232Th-rich materials, case study: town of Bolsena (Italy) entirely built with tuff
- Thoron concentration was found to decrease with a relaxation length of 13 cm.
- The rate of air exchange is irrelevant: ventilation cannot ensure thoron mitigation.
- Wall plaster (HVL of 0.95 cm) can contribute efficiently to thoron mitigation.

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ABSTRACT

The contribution of ^{220}Rn is usually negligible compared to that of ^{222}Rn: its very short half-life makes escape from its source site within the rock very unlikely and it never has time enough to filtrate through the ground and through cracks in the floors or cellar walls to reach living quarters. This however becomes untrue if walls built with ^{232}Th-rich materials are present: then sizeable amounts of thoron may be detected in the closed areas bounded by those walls. This is the case for many dwellings in central Italy, and the town of Bolsena (northern Latium) is presented as a case study. A typical building of the area, entirely constructed with tuff blocks, is investigated and the annual dose rates calculated for varying distances from the wall. Thoron concentration was found to decrease with a relaxation length of 13 cm. Thoron was found to pose a significant risk. The rate of air exchange was found to produce little effect. Wall plastering acts as a filter: thoron diffuses through it but a HVL of less than 1 cm was found to prevail.

1. Introduction

^{222}Rn-mostly referred to simply as radon-and its short-lived decay products in the atmosphere are the major contributors to human exposure from natural sources, producing an average effective dose to the population of 1.15 mSv/y (UNSCEAR, 2008). The average annual effective dose due to ^{220}Rn-usually referred to as thoron, the radon isotope belonging to the decay chain of ^{232}Th—and its progeny is estimated to be over 10 times smaller than that due to radon: 0.1 mSv/y (UNSCEAR, 2008). For this reason, exposure to thoron and its decay products has often been neglected in the past. However, several studies have shown that in some cases doses from thoron and its progeny can be comparable to those from radon and its short-lived decay products, or even larger (Steinhäusler, 1996; Porstendorfer, 1994; Khokhar et al., 2008).

The half-life plays a key role in determining people exposures: since the half-life of thoron ($t_{1/2}$=55 s) is very much shorter than that of radon ($t_{1/2}$=3.82 days), exposure modes are quite different, the distance that thoron can travel before undergoing radioactive decay is much shorter than that traveled by radon in the same medium. For these reason, the impact of thoron concentrations on exposure is restricted to some vicinity of the source, decreasing rapidly with distance from the latter (UNSCEAR, 2006). Due to these characteristics, an appropriate setting to investigate thoron behavior is found inside buildings constructed with thorium rich materials. The present paper reports the results of an experimental study conducted in Bolsena—a town of northern Latium (Italy)

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continuously settled since Etruscan times (probably as far as the 6th century BC)—in the framework of a wider research project concerning several radiological and geological aspects of this high background region (Cinelli, 2012; Cinelli et al., 2014; Capaccioni et al., 2012). The selection of this town was based on the widespread use of volcanic rocks pertaining to the Vulsini Volcanic District as building materials, mainly pyroclastic rocks with variable amounts of lithified ash matrix (ash and lapilli tuffs) which supports pumiceous lapilli of phonolitic and/or trachitic composition (Nappi et al., 1995; Capaccioni et al., 2012). Vulsini volcanic products are characterized by an uncommonly strong natural radiation background resulting from the high concentrations of Uranium, Thorium and Potassium (Capaccioni et al., 2012). This material has been used since the town's Etruscan times and is still found also in modern buildings, since the gravel in the concrete used in present day construction work comes from local quarries of the same volcanic stone. In particular, downtown Bolsena is entirely built in this local tuff stone. This state of affairs makes this town a prime location for investigating thoron.

2. Material and methods

2.1. The Vulsini volcanic district: Bolsena complex

The Vulsini Volcanic District (Nappi et al., 1995; Capaccioni et al., 2012), which was active in the period 570–127 ka (Gillot et al., 1991), is located in the northern part of the Quaternary potassic volcanic belt of the Roman Magmatic province (central-southern Italy). The Vulsini volcanic District consists of hundredth of eruptive centres and at least four collapsed structures ("Volcanic calderas"). These were grouped into four volcanic complexes: Paleo–Bolsena, Bolsena, Latera and Montefiascone (Nappi et al. 1991). The development of each complex was based on alternating effusive and explosive eruptions, with Strombolian, Plinian and Ignimbrite-forming phases. Processes of magma genesis and evolution produced a marked enrichment of radionuclides in the potassic magmas emitted during the quaternary age in the Vulsini Volcanic District. In this context, subduction-related metasomatic enrichment of incompatible elements in the mantle source coupled with magma differentiation within the upper crust has given rise to melts particularly enriched in U, Th and K.

2.2. High resolution γ spectrometry

The activity concentrations of ^{228}Ac had been measured in twenty-one rock samples through high resolution γ-ray spectrometry and the concentration of the parent element ^{232}Th calculated (Capaccioni et al., 2013). The values ranged between 126 and 487 Bq/kg, a concentration level well above the average reported in international literature: 7–80 Bq/kg (Paschoa and Steinhäuscer, 2010). In the present work the thoron emitting walls have been assumed built in Sorano tuff, for which a ^{232}Th activity of 322 Bq/kg had been determined (Capaccioni et al., 2012). This rock was chosen as one of the most widely used building materials in the area.

2.3. In situ thoron measurements (RAD7)

Thoron concentration was measured using a RAD-7 radon monitor (Durridge Co.). Briefly, the instrument works by drawing air and circulating it through a membrane filter to retain airborne particulate matter containing radon and thoron progeny after passing it through a desiccant trap (drierite) to prevent artifacts during measurements. Purified air from soil interstices is hence drawn inside the RAD-7 measuring chamber where thoron gas is detected through α-decay of its daughter ^{216}Po produced therein. The instrument was operated in Sniff mode (RAD7, 2009). The short half-lives of thoron (55 s) and of its first decay product (Po-216-0.15 s) imply that thoron measurements can be made quickly and in rapid succession-the RAD7 responds virtually instantly: its time constant for response to thoron is less than 1 min. In each point more than one measurement was taken and the final result was estimated as the average thereof.

Table 1
RESRAD-BUILD input parameters

Parameter	Value
Thoron emanation coefficient	0.2
Occupancy factor	0.8
Air exchange rate [h^{-1}]	0.5
Wall erosion [cm/d]	2.4×10^{-8}
Air release fraction	0.1
Wall porosity	0.2
Thoron diffusion coefficient [m^2/s] (Nazaroff and Nero, 1988)	1×10^{-7} for concrete 1×10^{-6} for plaster

2.4. RESRAD-BUILD

In the present work, the internal dose due to inhalation of aerosol indoor thoron progeny in a model room have been evaluated by using the RESidul RADioactivity in BUILDings (RESRAD-BUILD) code (DOE, 2003). It is assumed that the building materials, with a known content of ^{232}Th, are the only source of thoron. The input parameters are summarized in Table 1.

The code uses dose conversion factors for inhalation taken from the EPA's (US Environmental Protection Agency's) federal guidance report no. 11 (FGR11, Eckerman et al., 1988). The RESRAD-BUILD has no transport capabilities, and can calculate no spatial dependence: it yields the average concentration in the room, seen as homogenous. It takes into account ventilation, so the dose calculated is, in general, a function of the air exchange rate. The following two possible remediation practices have been investigated: (a) the effect of varying air exchange rate, and (b) the influence of wall plastering.

3. Results and discussion

3.1. Experimental relaxation length

Thoron concentration was measured at different distances from the walls (3, 10, 15, 20 and 40 cm) to assess migration distance from the wall surface. The measurements have been taken indoor, in a public building of Bolsena entirely built with tuff blocks composed by mixed trachitic welded and trachiphonolitic secondary lithified tuffs ("sillar"; Capaccioni et al., 2001). In Table 2 the results of all measurements are reported, together with the averages at the measurement points.

As expected, thoron concentration decreases steeply with distance from the wall, ranging from 1570 Bq/m^3 at 3 cm to 369 Bq/m^3 at 20 cm. From the experimental data in Table 2 the relaxation length has been estimated to be 12.7 cm. In literature, values around 4 cm are reported for open air concentrations (Nazaroff and Nero, 1988; Meisenberg and Tschierch, 2010).

However, due to the large ventilation rate thoron concentration in open air is not expected to decrease exponentially with distance from the source, so that distribution may be more complicated (Urosević et al. 2008). Some reports for indoor diffusion are found in literature, see e.g. Ujić et al. (2010) who predicted a relaxation length of 20 cm. In the present case the measurements were

Table 2
Indoor thoron concentration measurements (Bq/m^3) at varying distances from the wall and average values thereof.

Distance from wall [cm]	Single measurement [Bq/m^3]	Average concentration [Bq/m^3]
0	2110 ± 844 1990 ± 824 1570 ± 747	**1865 ± 463**
3	1150 ± 660 1630 ± 759	**1357 ± 498**
10	603 ± 520 904 ± 603	**731 ± 394**
15	600 ± 518 663 ± 538	**630 ± 373**
20	600 ± 538 300 ± 416 300 ± 416	**369 ± 258**

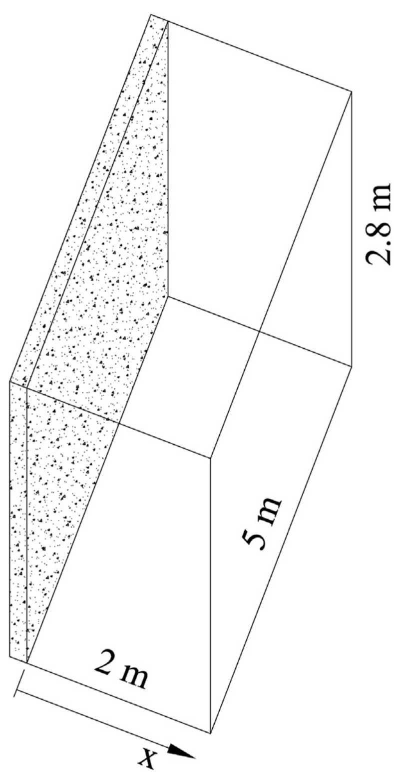

Fig. 1. Geometry of the simulation.

conducted in a real building with all the wall made in tufaceous blocks and with normal everyday ventilation present, so that the mix of convection and diffusion can be regarded as representative of everyday local conditions.

3.2. Dose estimation and its variation with distance

Numerous investigations have been conducted to simulate indoor behavior of radon and thoron and their progenies with the purpose of estimating the health risk (e.g., Urosević et al. 2008; Meisenberg and Tschierch, 2010; Ujić et al., 2010). As pointed out by numerous authors (see e.g., Meisenberg and Tschierch, 2010; Ujić et al., 2010), whereas the first daughter of toron, ^{212}Po, is very

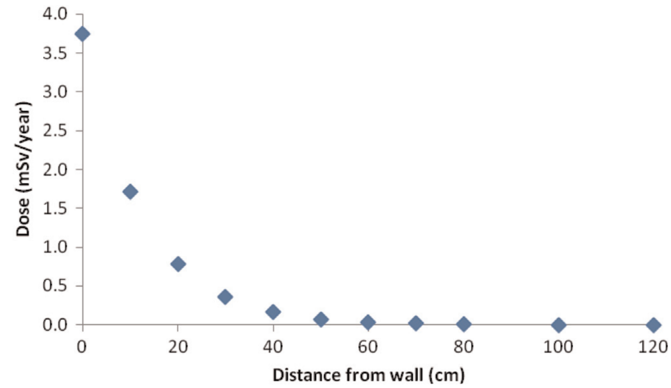

Fig. 2. Dose in mSv/year at different distance from the wall in the simulation room.

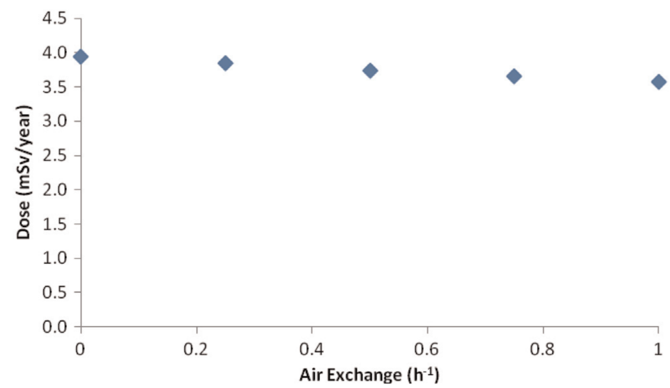

Fig. 3. Dose at the wall in the model room for varying air exchange values.

short lived (half-life 0.145 s), the following one, ^{212}Pb, with its comparatively long half-life (10.64 h), and has time to diffuse throughout the room, so that it can be considered essentially homogeneously distributed over the volume. On the contrary, thoron with its rather short half life (55.6 s) is confined to the vicinity of the emitting surface. This makes for a mixed problem, further altered by convective motions always present in a real setting. In the present work the method applied to estimate the dose attempts to strike a compromise between the uniform spatial distribution of thoron progeny-RESRAD-BUILD calculates the average concentration of thoron and its progeny in the room and from this the average dose due inhalation-and the exponential spatial distribution of thoron with the relaxation length determined experimentally. To this end, the following room was simulated, representing the half room relevant to the wall examined: only the wall of interest contained the specified concentration of ^{232}Th and its progeny (discussed in 2.2); the remaining surfaces (walls, ceiling, floor) contained none and served only the purpose of enclosing the volume: the geometry is shown in Fig. 1. This particular configuration was selected to model the fact that albeit the code assumes uniform distribution, in reality no thoron can reach from one surface to another.

The dose variation with distance from the wall (the only source) has been estimated assuming exponential decay with the relaxation length discussed above and equating the average of the exponential curve to the value yielded by RESRAD-BUILD, according to the following equation:

$$D(x) = D_R \frac{\exp\{-x/\lambda\}}{\frac{1}{2}\int_0^2 \exp\{-x/\lambda\}\mathrm{d}z} \tag{1}$$

where, D_R is the average yearly dose (mSv/year) in the model room estimated by RESRAD-BUILD (in the geometry described above), D

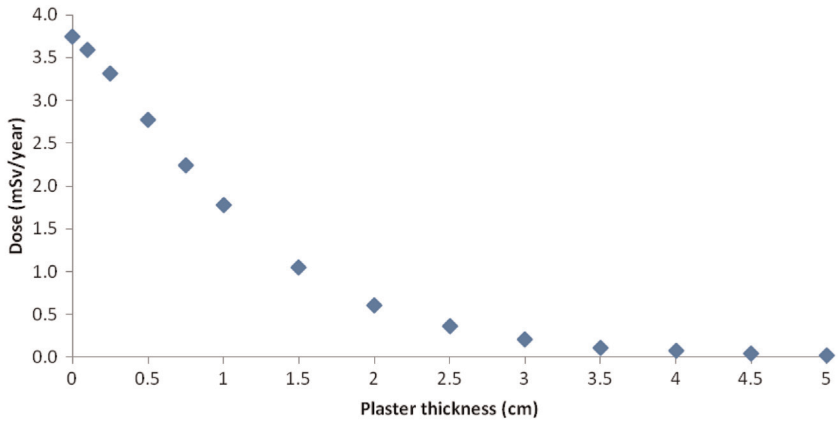

Fig. 4. Dose at the wall in the model room for varying thicknesses of plaster.

(x): the yearly dose (mSv/year) at a distance x from the wall, and λ is the relation length calculated in the present work (12.7 cm).

The dose variation with distance is reported in Fig. 2. The dose in contact with the wall has been estimated to 3.75 mSv/year and this value halves after about 8 cm. From Fig. 2 it can be noticed that at distances >50 cm, the dose is negligible, due to the sharp exponential decrease.

3.3. Air exchange

Ventilation efficacy in radon mitigation is well recognized, and its effect in connection with thoron and the dose from thoron progeny inhalation has also been investigated in the present work. Using the RESRAD-BUILD code the dose has been estimated in the model room (Fig. 1), at the wall contained the specified concentration of ^{232}Th and its progeny (see paragraph 2.2).

Ventilation appears to have no effect on thoron dose rate, at least at the ventilation rates considered, which are typical of real life buildings. This is easily explained considering the half-life of thoron: in a closed box equilibrium would be reached within about 5–6 min, hence even with 1 volume/h (excellent ventilation) only 1/10 of the air is exchanged in the time it takes thoron to build up.

Fig. 3 shows the dose estimated varying the air exchange values from 0 to 1 h^{-1}, this latter being considered typical of Italian buildings, with the usual window fixtures utilized in the Country.

3.4. Effect of plastering

As means of mitigation might be plastering the walls, which are usually exposed since the stone is considered aesthetically pleasing. Wall plaster (usually not containing significant amount of Th-232 and progeny) acts as a filter. The influence of plaster on the wall has been investigated.

Fig. 4 reports the dose at the wall containing the specified concentration of ^{232}Th and its progeny (see paragraph 2.2), in the model room, estimated using RESRAD-BUILD code: plaster thicknesses varying from 0 to 5 cm were considered. It can be seen that the half-value layer (HVL) is of the order of 0.95 cm.

4. Conclusions

Thoron was found to pose a significant risk. One striking feature of this hazard is that the rate of air exchange is irrelevant, contrary to what is the case with radon: hence ventilation cannot ensure mitigation. Wall plaster acts as a shield: thoron diffuses through with a HVL of 0.95 cm showing that it can contribute efficiently to dose reduction. Measurements at varying distances from wall surfaces showed a relaxation length of the order of 13 cm. Considering a building material with a content in Th-232 of 322 Bq/kg the distance at which no extra dose from thoron is received is about 40 – 50 cm. Considering 0.8 for the indoor occupancy factor an annual effective dose of about 4 mSv is calculated in close proximity (contact) with the wall.

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